

REMARKS

Claims 1 and 3-16, 18 and 19 are pending in the present application. Claims 16, 18 and 19 are withdrawn from consideration. Claim 1 is herein amended. No new matter has been presented.

Support for the amendments to claim 1 is at, *e.g.*, page 13, lines 9-18 and Examples 1-3, 5 and 6 of the specification.

Claim Rejections - 35 U.S.C. § 112

Claims 1 and 3-15 were rejected under 35 U.S.C. § 112, first paragraph, as failing to comply with the written description requirement.

The Office Action takes the position that the specification does not support the range of selectivity. (Office Action, page 3.) Claim 1 has been amended to remove the noted limitation.

Withdrawal of the § 112 rejection is requested.

Claim Rejections - 35 U.S.C. § 103

A. Rejection based on Wu

Claims 1 and 3-15 were rejected under 35 U.S.C. § 103(a) as being unpatentable over **Wu** (US 5,898,014). Favorable reconsideration is requested.

(1) Applicants respectfully submit that Wu does not teach or suggest:

A catalyst for manufacturing synthesis gas containing carbon monoxide and hydrogen as principal ingredients from feedstock gas containing hydrocarbon having 1 to 5 carbon atoms in each molecule and oxygen, characterized in that

the catalyst for manufacturing synthesis gas has an aluminum-free carrier and a catalyst metal carried by the carrier; [and]

said catalyst metal comprising at least a Group VIII metal including at least rhodium;

as recited in amended claim 1.

The catalyst composition of Wu comprises a support, a precious metal component and a zirconium, rare earth composition. (See col. 8, lines 45-48.) The zirconium, rare earth composition functions as oxygen storage components and constitutes the catalyst composition together with the support carrying the precious metal. The zirconium, rare earth composition as oxygen storage components is preferably in ‘bulk’ form, i.e., generally existing as a uniform composition having substantially no internal structure. (See col. 10, lines 17-21.) The support is in the form of particles having a diameter of less than 25 microns and comprises one or more refractory oxides having a high surface area, particularly including alumina as primary component. (See col. 9, lines 7-8 and 20-30.) Thus, the catalyst of Wu comprises the support particles carrying the precious metal thereon dispersed in a medium of the uniform composition of oxygen storage components (i.e. the zirconium or rare earth composition).

In contrast, the catalyst composition of the present invention comprises a carrier and a Group VIII element. The carrier in the present invention does not correspond to the support in Wu, but rather corresponds to the zirconium, rare earth composition in Wu. Thus, in the catalyst composition of the present invention, the Group VIII element as catalyst metal is carried uniformly by the carrier in ‘bulk’ form, while no “support” as in the catalyst composition of Wu exists. In other words, the carrier (hence the catalyst composition) in the present invention does not contain alumina which is the primary component of the support in Wu.

The Office Action takes the position that “a catalyst which can achieve oxidation can be used for a process of partial oxidation if the reactants are given in an amount which only allows for such, for example a less than stoichiometric amount of oxygen.” (Office Action, page 5.) However, partial oxidation cannot be practically achieved by only adjusting the proportion of reactants to be fed into the reaction furnace. To the contrary conditions must be optimized for partial oxidation. Two articles are attached which explain that partial oxidation of methane to synthetics gas (“syngas”) has been long investigated and it has been found that it involves a number of difficulties. See particularly Article 1, page 54, left column, lines 4-15 and Article 2, page 177, left column, lines 7-16.

Ideally, partial oxidation is carried out at O₂:CH₄=1:2 (for CH₄+1/2O₂ →CO+2H₂). If the oxygen content is higher than the ratio, CO and H₂ once formed (even under the ideal progress of reaction) will be combusted to produce increased amounts of CO₂ and H₂O and lower the methane conversion rate. On the other hand, if oxygen content is lower than the ratio, the selectivity for CO and H₂ may be unchanged or even improved. However, even if the selectivity for CO and H₂ is improved, since the amount of oxygen necessary for the reaction is absolutely insufficient, the methane conversion rate will be lowered. Thus, even if the oxygen content is lowered to improve the selectivity, the production efficiency of synthesis gas will be lowered. In other words, a catalyst for partial oxidation must show a high methane conversion rate and a high selectivity for CO and H₂ under the above ideal ratio of O₂:CH₄=1:2. In the examples of the present application, the ratio of O₂:CH₄:Ar=15:30:55 is employed. Comparative Example 15 in the present specification demonstrates that a catalyst substantially corresponding to the catalyst

of Wu ($\text{CeO}_2:\text{ZrO}_2=50:50$) shows a lower performance than the catalysts having a composition within the claimed range.

Wu discloses a catalyst for treatment of exhaust gas from automobiles. (Col. 1, lines 19-30.) In the automobile exhaust gas treatment intended in Wu, it is required to carry out in parallel three reactions including complete oxidation of hydrocarbon, complete oxidation of carbon monoxide to carbon dioxide, and complete reduction of nitrogen oxides to nitrogen, i.e. carrying out in parallel three different reactions up to completion. In contrast, in the catalytic partial oxidation of natural gas intended in the present invention, it is required to stop oxidation of natural gas (to carbon dioxide and water finally) at the stage where natural gas (predominantly methane) is converted to carbon monoxide and hydrogen, i.e. carrying out a single reaction so as to stop at a specific point in the midway.

In addition, the Declaration submitted June 22, 2009 establishes that a catalyst substantially equivalent to the catalyst in Wu is not a catalyst for manufacturing synthesis gas since as understood by one of ordinary skill in the art, the selectivity for CO and H₂ and conversion of methane are not satisfied for producing synthesis gas.

(2) Applicants respectfully submit that Wu does not teach or suggest:

wherein the molar ratio of said second ingredient relative to said first ingredient is between 0.10 and 0.25 and the molar ratio of said third ingredient relative to said first ingredient is between 0.3 and 0.6 as recited in amended claim 1.

Wu contains three examples, of which Examples 1 and 2 illustrate typical examples of the zirconium, rare earth composition (i.e. oxygen storage components) while Example 3

illustrates a catalyst composition containing the oxygen storage components of Example 1. The zirconium, rare earth compositions of Examples 1 and 2 comprise Zr, Ce, Pr and Nd, and does not contain an alkaline earth metal oxide (first ingredient) of the present invention. On the other hand, the catalyst composition of Example 3 contains, in addition to the zirconium, rare earth composition as oxygen storage components, Pd as catalyst metal carried on an alumina support, Sr as stabilizer, La and Nd as promoter, and Zr as additional oxygen storage component. This catalyst composition is formed as a coating layer on a ceramic honeycomb.

In Example 3 of Wu, the catalyst composition is formed on a ceramic honeycomb substrate by dipping the ceramic honeycomb substrate into a slurry containing the zirconium, rare earth composition (containing oxides of Zr, Ce, Pr and Nd) as oxygen storage components of Example 1, alumina support particles containing Pd (palladium nitrate) as catalyst metal, as well as Zr (zirconium acetate), La (lanthanum nitrate), Sr (strontium hydroxide) and Nd (neodymium nitrate) as additional components, then air knifing excessive coatings of the slurry to clear out the channels, drying the coated substrate, and calcining it at 550°C.

The catalyst composition formed on the ceramic honeycomb contains the following elements (except the elements constituting the alumina support particles):

Pd:	0.0868 g/in ³ (as Pd);
Zr:	0.100 g/in ³ (as ZrO ₂);
La:	0.190 g/in ³ (as La ₂ O ₃);
Sr:	0.100 g/in ³ (as SrO);
Nd:	0.160 g/in ³ (as Nd); and

as ingredients of the oxygen storage components of Example 1,

Ce:	0.500 g/in ³ x 0.28 (as CeO ₂);
Pr:	0.500 g/in ³ x 0.07 (as Pr ₆ O ₁₁);

Nd: 0.500 g/in³ x 0.07 (as Nd₂O₃); and
Zr: 0.500 g/in³ x 0.58 (as ZrO₂).

The atomic or formula weight of each ingredient is as follows:

Pd	=	106.42,
Zr	=	91.22,
La	=	138.91
(La ₂	=	277.82),
Sr	=	87.62,
Nd	=	144.24
(Nd ₂	=	288.48),
Ce	=	140.12,
Pr	=	140.91
(Pr ₆	=	845.46),
O	=	16.00,

hence,

ZrO ₂	=	91.22 + 16x2 = 123.22,
La ₂ O ₃	=	138.91x2 + 16x3 = 325.82,
SrO	=	87.62 + 16 = 103.62,
CeO ₂	=	140.12 + 16x2 = 172.12,
Pr ₆ O ₁₁	=	140.91x6 + 16x11 = 1021.46,
Nd ₂ O ₃	=	144.24x2 + 16x3 = 336.48.

Accordingly, the concentration in g/in³ and molar concentration in mol/in³ of each ingredient (except the alumina support particles) are as follows:

Pd:	0.0868 g/in ³ ;
SrO:	0.100 g/in ³ ; (0.100/103.62 = 0.965x10 ⁻³ mol/in ³);
La ₂ O ₃ :	0.190 g/in ³ ; (0.190/325.82 = 0.583x10 ⁻³ mol/in ³);
CeO ₂ :	0.500x0.28 = 0.14 g/in ³ ; (0.14/172.12 = 0.813x10 ⁻³ mol/in ³);
Pr ₆ O ₁₁ :	0.500x0.07 = 0.035 g/in ³ ; (0.035/1021.46 = 0.034x10 ⁻³ mol/in ³);
Nd ₂ O ₃ :	0.160x336.48/288.48 + 0.500x0.07 = 0.222 g/in ³ ; (0.222/336.48 = 0.659x10 ⁻³ mol/in ³);
ZrO ₂ :	0.100 + 0.500x0.58 = 0.39 g/in ³ ;

($0.39/123.22 = 3.16 \times 10^{-3}$ mol/in³).

Of the above ingredients, corresponding to the first ingredient is SrO, to the second ingredient are La₂O₃, CeO₂, Pr₆O₁₁ and Nd₂O₃, and to the third ingredient is ZrO₂. Therefore,

$$\text{2nd/1st molar ratio} = \frac{0.583 + 0.813 + 0.034 + 0.659}{0.965} = 2.16, \text{ and}$$

$$\text{3rd/1st molar ratio} = \frac{3.16}{0.965} = 3.28.$$

As seen from the above, the catalyst composition (except alumina support particles) described in Example 3 of Wu has molar ratios significantly out of the range for molar ratios of the present invention (i.e. 2nd/1st = 0.02-0.40 and 3rd/1st = 0.04-1.5 as recited in claim 1). Roughly, the content of the first ingredient in the catalyst composition of Wu is small as compared with the catalyst composition of the present invention.

Wu mentions tolerable or preferable ranges for the contents of the individual elements (Zr, Ce, Nd and Pr) constituting the oxygen storage components, but Wu does not mention any ranges for the content ratios among these elements as acknowledged in the Office Action at page 5. Claim 25 of Wu recites preferable ranges for the content of oxygen storage components, the contents of Zr and rare earth metals as additional oxygen storage component or promoter, and the content of alkaline earth metals as stabilizer, with considerable breadth, but it does not recite any ranges for the content ratios among those ingredients.

When the contents of individual elements are defined with ranges of considerable breadth, it is unreasonable to derive a range for the content ratios among those elements from the ranges for the contents of the individual elements. While the Office Action calculated ranges for the

content ratios among elements from the ranges for the individual contents of those elements, the Office Action's procedure of calculation is not proper. The unreasonableness of this calculation can be seen from the very broad ranges resulting from the calculation. (Office Action, page 6 calculating the following range of ratios: 0.0126 to 18.73 and 0.0189 to 32.71.)

As noted above, based on the specific example in Wu (Example 3), since the molar ratios of second to first ingredients and of third to first ingredients calculated from Example 3 of Wu are significantly out of the ranges defined in the present invention, the recited range of ratios for the ingredients would not have been obvious based on the disclosure in Wu.

(3) Applicants respectfully submit that Wu does not teach or suggest "an aluminum-free carrier" and that the catalyst metal comprises "at least a Group VIII metal including at least rhodium."

Contrary to Wu, the catalyst composition of the present invention does not contain alumina because of the fundamental difference attributable to the difference in the technical field and the problem to be solved.

While the catalyst composition of Wu contains alkaline earth metals as stabilizer (for alumina support particles) and typically Sr is used as seen in Example 3, the catalyst of the present invention contains alkaline earth metals just as carrier component and primarily Mg is used as seen in Examples 1-3 and 5-6. This is apparently because the catalyst composition of Wu is intended to be used as a catalyst for automobile exhaust gas in a heated state at 1000°C or higher and the alumina (gamma-alumina) support material can possibly cause phase transfer, whereas the catalyst composition of the present invention is intended to be used as a catalyst for

partial oxidation of natural gas to produce synthesis gas in a state controlled so as to be not heated at 1000°C or higher. Since the catalyst composition of the present invention does not contain alumina, there is no need for using a stabilizer for alumina.

Furthermore, the catalyst composition of Wu contains a precious metal as catalyst metal which is typically platinum or palladium, while the catalyst of the present invention contains primarily rhodium. Rhodium is not a ‘platinum group metal’ and has more or less different properties from platinum group metals in view of the disclosure of Wu at col. 2, lines 9-11.

In addition, while the zirconium, rare earth composition as oxygen storage components constituting the catalyst composition of Wu contains Zr, Ce, Nd and Pr as essential elements, the catalyst composition of the present invention does not necessarily contain these four elements. In the present invention, while Zr and Ce are essential as the third and second ingredients respectively, Nd and Pr are not essential.

B. Rejection based on Niu, Yagi and Allison

Claims 1 and 3-15 were rejected under 35 U.S.C. § 103(a) as being unpatentable over **Niu** (US 2003/0180215) in view of **Yagi** (US 6,376,423) and **Allison** (US 2002/0115730). Favorable reconsideration is requested.

Applicants respectfully submit that the present invention as recited in the claims provides unexpected results and thus, the molar ratios recited in the claims are critical. The results from the Examples and Comparative Examples as summarized in Tables 1 and 2, demonstrate that when the molar ratio of the second ingredient to the first ingredient and the molar ratio of the third ingredient to the first ingredient go out of their respective ranges, the conversion rate and

the selectivity fall. (Specification, page 27.) The results also demonstrate that when the ratio of the third ingredient to the first ingredient increases, the CO and H₂ selectivity increase, but the methane conversion rate and the resistance against carbon deposition are not satisfactory. (Specification, page 27.) Additionally, the results demonstrate that when the ratio of the second ingredient to the first ingredient increases, the methane conversion is improved, but the CO and H₂ selectivity decrease and the resistance against carbon deposition is not satisfactory. (Specification, pages 27-28.) Therefore, the results in Tables 1 and 2 demonstrate unexpected results and the criticality of the recited molar ratios of the carrier ingredients.

For at least the foregoing reasons, claim 1 is patentable over the cited references, and claims 3-15 are patentable by virtue of their dependence from claim 1. Accordingly, withdrawal of the rejection of claims 1 and 3-15 is hereby solicited.

In view of the aforementioned amendments and accompanying remarks, Applicants submit that the claims, as herein amended, are in condition for allowance. Applicants request such action at an early date.

If the Examiner believes that this application is not now in condition for allowance, the Examiner is requested to contact Applicants' undersigned attorney to arrange for an interview to expedite the disposition of this case.

Application No. 10/583,152
Art Unit: 1793

Amendment under 37 C.F.R. §1.114
Attorney Docket No. 062610

If this paper is not timely filed, Applicants respectfully petition for an appropriate extension of time. The fees for such an extension or any other fees that may be due with respect to this paper may be charged to Deposit Account No. 50-2866.

Respectfully submitted,

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Attachments: Article 1: K.L. Hohn et al., "Partial oxidation of methane to syngas at high space velocities over Rh-coated spheres", Applied Catalysis A: General 211 (2001) 53-68.

Article 2: R.S. Drago et al., "Partial oxidation of methane to syngas using NiO-supported catalyst", Catalysis Letters 51 (1998) 177-181.